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Improvement of Piezoelectricity in Piezoelectric Paper made with Cellulose

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ABSTRACT

This report deals with the improvement of piezoelectricity in the piezoelectric paper made with cellulose. Piezoelectric paper fabrication process is composed of cellulose dissolving, wet cellulose casting, regenerating cellulose, stretching and drying processes. To automate these serial processes, a pilot plant was developed for the wet cellulose casting, regenerating, stretching and drying processes. A belt casting system was developed so as to continually cast the wet cellulose film and regenerate it. A hot roller system was developed for zone stretching and drying of the wet cellulose film. In the cellulose regeneration process, ions and solvent were entirely eliminated by washing the cast cellulose film with deionized water/isopropyl alcohol mixture. The improvement of piezoelectric paper was investigated by measuring the mechanical properties and piezoelectric constant. The material properties of piezoelectric paper were greatly stabilized, and the piezoelectric constant was drastically improved, comparing with the manually made piezoelectric paper. To demonstrate its application possibility, the fabricated piezoelectric paper was applied to make paper speaker, vibration sensor and acoustic wave device.

Keywords: Piezoelectric paper, Cellulose, Mechanical stretching, Paper Speaker, Vibration Sensor, Electro-Active paper.

1. Introduction

Cellulose Electro-active paper (EAPap) has been recognized as a new smart material that can be used for sensors, MEMS devices, actuators, bio-mimetic robots, smart wall papers, and micro flying objects [1]. EAPap is made with cellulose paper by coating thin electrodes on both sides of it. This paper can produce a bending or longitudinal strain in the presence of electric field. Also, it produces an induced charge under the external stress. This EAPap material has many advantages in terms of large displacement output, low actuation voltage, low power consumption, dryness, low price, flexibleness, sensing capability and biodegradable characteristics.

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14. ABSTRACT
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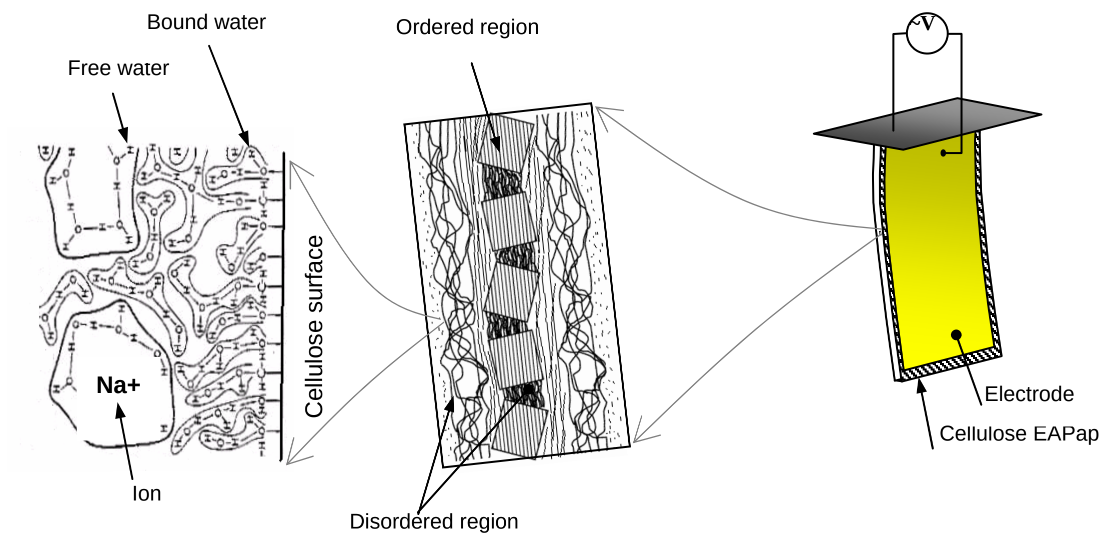
Based on the cellulose structure and the processing of the cellulose-based EAPap, it has been found that the actuation is due to a combination of two mechanisms: ion migration and piezoelectric effect [2]. Cellulose EAPap material is composed of molecular chains with a dipolar nature. In particular, the crystal structure of cellulose II is monoclinic, which is noncentro-symmetric and exhibits piezoelectric and pyroelectric properties. Structural changes of native cellulose crystals have been made by annealing in aqueous alkaline and acid solutions in high temperature. Aligning cellulose chains in paper is very important to improve piezoelectricity in cellulose paper.



c. b. a.

Figure 1 Concept of Electro-Active Paper Actuator: **a.** Cellulose microfibril has ordered crystalline regions and disordered regions. **b.** EAPap is made from cellulose paper on which gold electrodes are deposited on both sides. **c.** water molecules are bonded with hydroxyls on the cellulose surface (bound water) or clustered in free (free water) [2].

The piezoelectricity gives a great potential of EAPap as biomimetic actuators and sensors. According to our early investigation of piezoelectricity in EAPap, it showed a remarkable piezoelectric constant. Piezoelectricity of EAPap is strongly dependent on the material orientation of cellulose EAPap and alignment of disordered regions in cellulose. When the material orientation was investigated, 45° orientation angle exhibited the piezoelectric constant of 28.2 pC/N, which is larger than well-known PVDF piezoelectric polymer. This 45° orientation angle might be strongly associated with crystal structure of cellulose that exhibits high piezoelectricity. We termed this cellulose EAPap that shows high piezoelectricity as ‘Piezoelectric Paper’.

However, the properties of piezoelectric paper have shown a wide variation depending on the fabrication process. So far, we have fabricated the piezoelectric paper by using separated processes: Cellulose solution fabrication, tape casting, washing with deionized (DI) water/isopropyl alcohol (IPA) mixture, stretching and drying. To eliminate the fabrication errors and improve the properties, a pilot plant was developed.

Also, to demonstrate the advantages of piezoelectric paper, it was applied for making paper speaker, vibration sensor and acoustic device.

2. Experimental

2.1 Pilot plant for piezoelectric paper fabrication

Figure 2 shows the schematic of pilot plant. This process consists of cellulose solution fabrication, casting of cellulose solution onto belt casting system, regenerating of cellulose by washing with DI water/IPA mixture, stretching and drying process. In the cellulose solution process, cotton pulp (MVE, DPw 4580) purchased from Buckeye Technologies Co., USA and LiCl (Junsei Chemical) were heated in an oven at 100℃ to evaporate water. The cotton pulp was mixed with LiCl/anhydrous DMAc (*N,N*-dimethyl acetamide) (Aldrich). The cellulose was dissolved in the solvent by heating at 155℃ with mechanical stirring according to the solvent exchange technique [3]. The cellulose solution was coated on a belt system using a doctor blade. It was cured in the solvent mixture, which is composed of DI water and IPA, for 3 hours to effectively eliminate Li^+ ions as well as DMAc [4]. The cured wet cellulose film was sent to hot roller system that is supposed to zone stretch and dry. Hot rollers dry the wet cellulose film. By slightly increasing the hot roller speed from the entrance to exit, a continuous stretching can be made between rollers. This is so called zone stretching. The final dried sample was wound on a reel system.

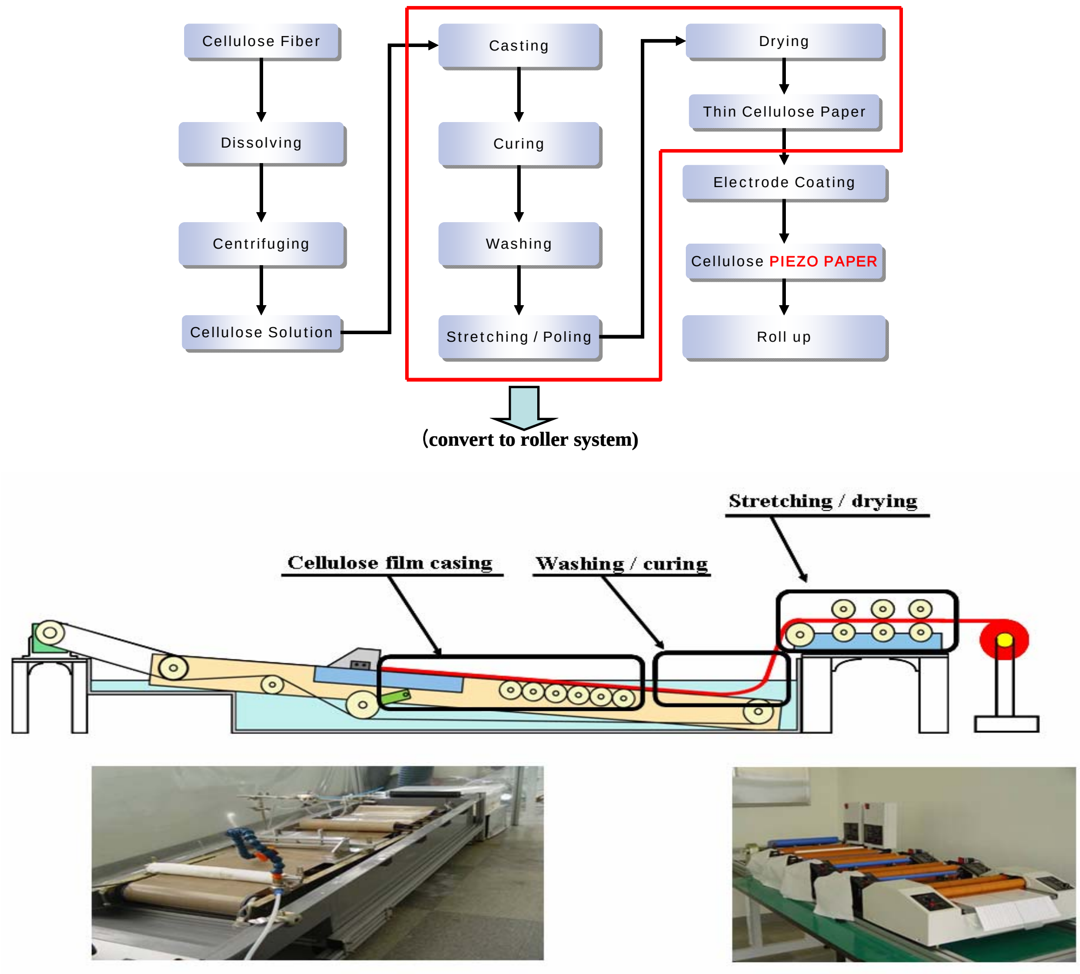


Figure 2 Pilot plant for cellulose piezoelectric paper fabrication.

2.2 Material property characterization

The surface image of the fabricated piezoelectric paper was seen by SEM. Mechanical properties of the sample were measured using a pull test machine, which is composed of linear scaler (Sony, GB-BA/SR128-015), load cell (Daecell, UU-K010), automated linear motion controller, AD interface, computer and LabView. The pull test machine is located into an environmental chamber that can control temperature and humidity. To characterize the piezoelectricity, the induced charge of the cellulose EAPap was measured by quasi-static method. Cellulose piezoelectric paper on which gold electrodes were coated on both sides was installed in the pull test machine by gripping the sample with the ASTM standard grip for polymer film tests. The electrodes were wired to piccoammeter (Keithley, 6485) and LabView was used to acquire the displacement, force and induced charge data. The induced charge during the pull test was measured by the piccoammeter. The test temperature and humidity conditions were 24℃ and 20-25%RH. Once the induced charge is measured, in-plane piezoelectric charge constant can be found from the ANSI/IEEE standard [5].

2.3 Fabrication of application devices:

To demonstrate the piezoelectricity of cellulose EAPap, a paper speaker was made and tested. Issues in designing paper speaker are enough sound power and quality. Film speakers have made with PVDF piezoelectric polymer. Advantages of paper speaker above the film speaker are its biodegradability and low price. Target frequency range of paper speaker was 500-20,000 Hz. The low frequency band of paper speaker is limited due to the speaker size. Specialized amplifying electronic circuit for paper speakers was provided to support the conceptual demonstration of paper speaker.

Also, the possibility of vibration sensor of piezoelectric paper was demonstrated by attaching the piezoelectric paper sensor on a beam structure. To compare the sensing capability, an accelerometer was attached on the beam structure, and the frequency response functions (FRF) of the beam caught from the paper sensor and the accelerometer were compared. This test attempt will bring a new area of sensor applications.

Since piezoelectric paper can produce acoustic signals at high frequency, it can be used for structural health monitoring patch (SHMP). SHMP should be able to bond on a flexible UAV structure and remotely sense cracks in the structure. The concept of SHMP that we propose is based on an acoustic wave device. Interdigit transducer (IDT) made on cellulose piezoelectric paper can produce acoustic waves once electrical signal is applied onto it, and the waves propagate. As the waves hit cracks or defects on a structure where SHMP bonded like a sticker, the waves reflect back and they can inversely produce an electrical signal from the IDT. By attaching dipole microstrip antenna on IDT, remote activation and sensing can be possible. This patch is convenient for structural health monitoring. By scanning a microwave transducer that transmits electrical signal to SHMP and receives reflected signals from it, the structural characteristics can be detected. In this research, the fabrication of IDT pattern on piezoelectric paper and acoustical behavior were investigated using a network analyzer.

3. Results and Discussions

3.1 Characterization

Mechanical properties of the piezoelectric paper were evaluated in terms of Young's modulus and yield strength according to the orientation. The stretching ratio of the piezoelectric paper made by the pilot plant was fixed to 1.6. Young's modulus and yield strength of the pilot plant sample were 18.3 GPa and 140 MPa, respectively. These are very similar to the values for manually fabricated piezoelectric paper. Figure 3 shows the Young's modulus of the pilot plant sample depending on the orientation.

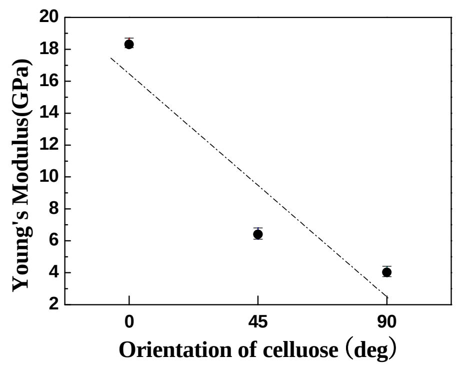


Figure 3 Young's modulus of piezoelectric paper.

Piezoelectric charge constant of the pilot plant samples was measured according to the method described before. Table 1 shows the results. Three samples for each orientation case were tested and the averaged. The pilot plant samples show larger d_{31} values than the manually process ones. Note that the maximum d_{31} was found when the orientation is 45° for the both manual and pilot plant cases. The maximum piezoelectric charge constant of the pilot plant sample was found to be 30.6 pC/N. This value is nearly three times higher than the manually made sample. This is larger than the PVDF piezoelectric polymer, 23 pC/N.

Table 1 Piezoelectric charge constant [d_{31}] with material orientation.

	Piezoelectric charge constant(pC/N)		
	Orientation of Material(cellulose)		
	0°	45°	90°
Manual process (SR=1.5)	5.9	10.7	1.9
Pilot plant process (SR=1.6)	22	30.6	10.7

3.2 Fabrication of application devices

Paper speaker was made by coating gold electrodes on both sides of the piezoelectric paper. Size of the electrode was 100 x 70 mm. To realize the potential piezoelectric performance of piezoelectric paper as a paper speaker, we measured its sound pressure level (SPL) in an anechoic chamber. Figure 4 shows the measured SPL the piezoelectric paper speaker with the input signal of 28 V. As shown, the maximum output of the piezoelectric paper speaker was 53 dB. The relatively low speaker performance below 5 kHz can be improved by optimizing the piezoelectric paper thickness and the design layout of the speaker. From the SPL result, we demonstrated the possibility of cellulose piezoelectric paper as an acoustic application.

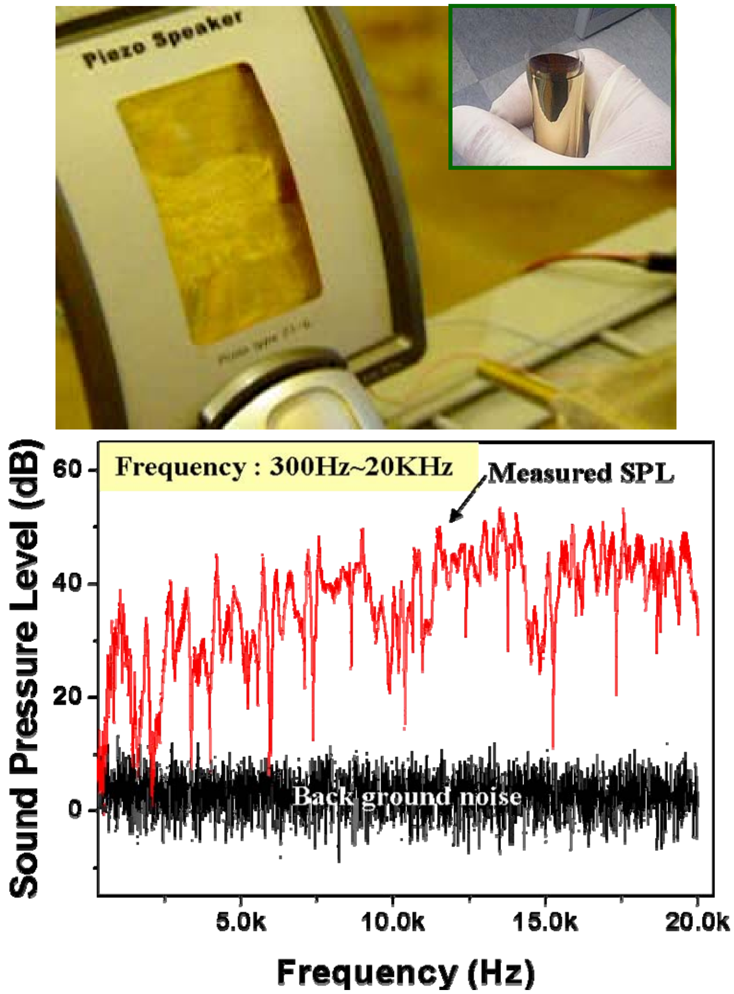
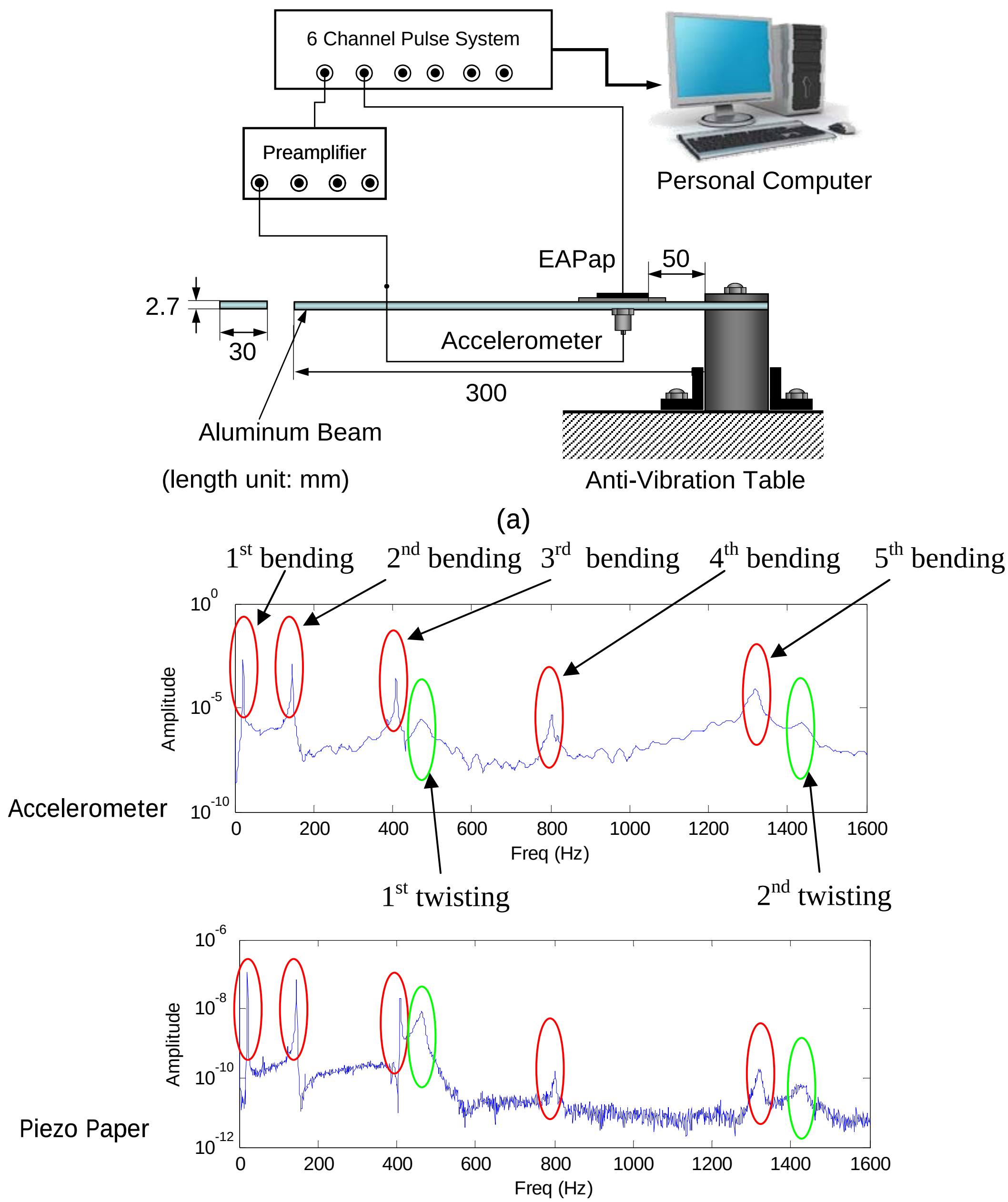


Figure 4 Paper speaker made with cellulose piezoelectric paper and its SPL.

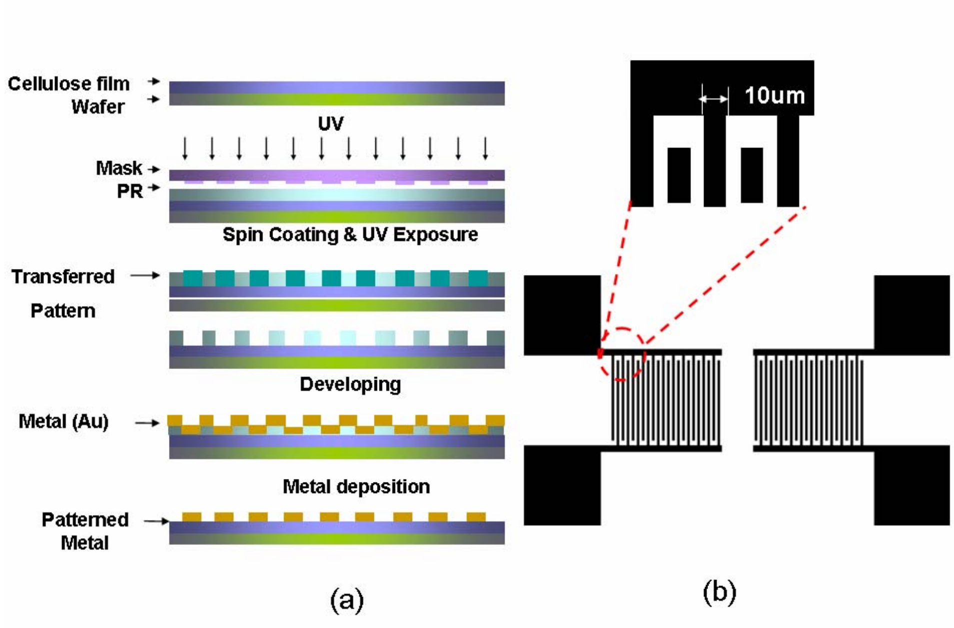
To test the vibration sensing capability of cellulose piezoelectric paper, the fabricated piezoelectric paper was attached on the aluminum beam as shown in Figure 5. Size of the beam was 30 cm length, 3 cm width and 2.7 mm thickness. The signal was captured by pulse analyzer (signal controller, 7536 and input module, 3039, B&K). For comparison, accelerometer (4393, B&K) is attached at the same place. The signal of accelerometer was amplified by the charge amplifier (NEXUS 2692, B&K) and data was acquired by the pulse analyzer. Time response of the cantilevered beam was obtained when impulsive load was applied to the beam by the impact hammer. Fast Fourier Transform (FFT) analysis was conducted to study frequency responses of the EAPap sensor. The impulsive response of piezoelectric paper provided correct dynamic characteristics of the beam. Especially, twisting mode of the beam was clearly observed even though the piezoelectric paper was attached at the center of the beam. This is because the sensing capability of piezoelectric paper is based on piezoelectricity which is bidirectional strain characteristics. From the observations, piezoelectric paper provided a great potential as a vibration sensor.

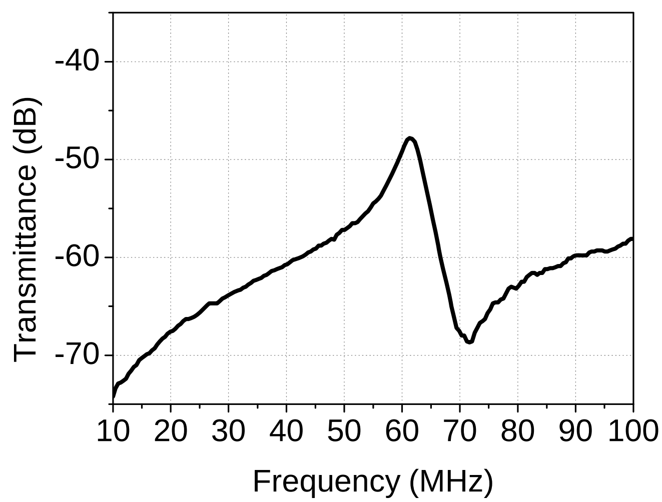


(b)

Figure 5 Vibration sensor test for piezoelectric paper: (a) Test setup (b) FRFs.

To investigate the concept of SHMP, an acoustic device was made with the piezoelectric paper. Figure 6 shows the fabrication of acoustic device made with cellulose piezoelectric paper. Interdigit transducer (IDT) made on the piezoelectric paper was made by lift off process (Fig. 6 a). Comb width of the IDT was made to be 10 μm under the assumption that the wave length, λ, is 40 μm. Number of finger pair is 60 and the aperture length is 50 λ and the acoustic path length is 150 λ (Fig. 6 b). The characteristic of the device was measured by using a network analyzer (Agilent N5230A). Transmission of the device was shown in Figure 6 (c) as a frequency function. From the transmission, the speed of sound of the piezoelectric paper was found to be 2452 m/s. Further acoustic characteristics of the device are under investigation.





(c)

Figure 6 Acoustic device made with piezoelectric paper: (a) lift off process, (b) schematic of IDT for the acoustic device, (c) frequency characteristic of the device.

4. Conclusions

In this research, improvement of piezoelectric paper was made by developing a pilot plant of piezoelectric paper fabrication. Mechanical properties of the pilot plant samples were similar with the manually made ones. Piezoelectric charge constant of the pilot plant samples were nearly three times higher than the manually made ones. To demonstrate its application possibility, paper speaker, vibration sensor and an acoustic device were made with the cellulose piezoelectric paper. It was shown that with the piezoelectric paper, acoustic signals up to ultra sound range can be generated and detected. Further investigation of the acoustic device is necessary for structural health monitoring application.

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